

# Emission Rates and Characterization of Aerosols Produced During the Spreading of Dewatered Class B Biosolids

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This study measured aerosol emission rates produced during the spreading of dewatered class B biosolids onto agricultural land. Rates were determined in multiple independent experimental runs by characterizing both the source aerosol plume geometry and aerosol concentrations of PM<sub>10</sub>, total bacteria, heterotrophic plate count bacteria (HPC), two types of biosolids indicator bacteria, endotoxin, and airborne biosolids regulated metals. These components were also measured in the bulk biosolids to allow for correlating bulk biosolids concentrations with aerosol emission rates and to produce reconstructed aerosol concentrations. The average emission rates and associated standard deviation for biosolids PM<sub>10</sub>, total bacteria, HPC, total coliforms, sulfite-reducing *Clostridia*, endotoxin, and total biosolids regulated metals were  $10.1 \pm 8.0$  (mg/s),  $1.98 \pm 1.41 \times 10^9$  (no./s),  $9.0 \pm 11.2 \times 10^7$  (CFU/s),  $4.9 \pm 2.2 \times 10^3$  (CFU/s),  $6.8 \pm 3.8 \times 10^3$  (CFU/s),  $2.1 \pm 1.8 \times 10^4$  (EU/s), and  $36.9 \pm 31.8$  (μg/s) respectively. Based on the land application rates of spreaders used in this study, an estimated  $7.6 \pm 6.3$  mg of biosolids were aerosolized for every 1 kg (dry weight) applied to land. Scanning electron microscopy particle size distribution analysis of the aerosols revealed that greater than 99% of the emitted particles were less than 10 μm and particle size distributions had geometric mean diameters and standard deviations near  $1.1 \pm 0.97$  μm. The demonstrated correlations of bulk biosolids concentrations with aerosol emission rates, and the reconstruction of aerosol concentration based on PM<sub>10</sub> and bulk biosolids concentration provide a more fundamental, bulk biosolids-based approach for extending biosolids aerosol exposure assessment to different land application scenarios and a broader range of toxins and pathogens.

## Introduction

Sewage sludges that have been stabilized to reduce the concentration of pathogens and indicator organisms, reduce

odors and putrefaction potential, and have a beneficial reuse are termed biosolids. In the United States, 60% of the 7 million dry tons of biosolids produced annually are reused by application to agricultural land (1). Land application provides a means of recycling the large quantities of residuals produced during municipal wastewater treatment, improves soil structure and water-holding capacity, and partially or fully replaces the use of conventional chemical fertilizer (2). However, biosolids contain pathogens and toxins, and persistent anecdotal allegations of negative health effects have emerged from citizens living near class B biosolids land application sites (3). An independent study by the National Research Council, National Academies of Science and Engineering, and the Institute of Medicine evaluated the technical methods and approaches used to establish the chemical and pathogen standards listed in the U.S.EPA biosolids land application guidelines. Relative to the topic of on-site and off-site human exposure, the report concluded that the inhalation pathway had not been adequately evaluated when the U.S.EPA set biosolids guidelines. While the required site restrictions during and after biosolids land application control the inhalation of dust on-site, U.S.EPA guidelines do not consider potential off-site exposure due to bioaerosols (4).

Quantitative assessment of acute and chronic health effects requires information on the concentration of both source and off-site biosolids-derived aerosols. An aerosol emission rate (number of microorganisms, mass of biotoxins, or mass of chemical compounds per time) is a required and highly sensitive input variable for all aerosol fate and transport models that predict absolute concentration at a specified distance. Emission rates also form an appropriate basis for comparing the amount of biosolids mass aerosolized between different biosolids types and land application processes. While previous indicator organism studies have clearly demonstrated that the aerosolization of biosolids occurs when dewatered biosolids (20–30% solids content) are loaded into application equipment or when liquid (2–8% solids content) and dewatered biosolids are spread onto land (5–7), limited data exist for emission rates (6). No published studies have reported measurement of aerosol emission rates for the common practice of land-applying dewatered biosolids, nor has information on critical human health parameters such as biosolids PM<sub>10</sub> concentrations, metals aerosol concentrations, or aerosol particle size distribution been detailed.

In response, bulk biosolids and the aerosols produced from the spreading of dewatered class B biosolids were characterized to determine source aerosol concentrations and aerosols emission rates for a suite of relevant chemical compounds and biological agents. To investigate an approach for simplifying biosolids bioaerosol assessment, PM<sub>10</sub> measurement and bulk biosolids concentrations were then used to demonstrate the reconstruction of source aerosol concentrations and correlate emission rates of specific bioaerosols or chemical compounds with the concentration of the same biological agents or chemical compounds in the bulk biosolids.

## Materials and Methods

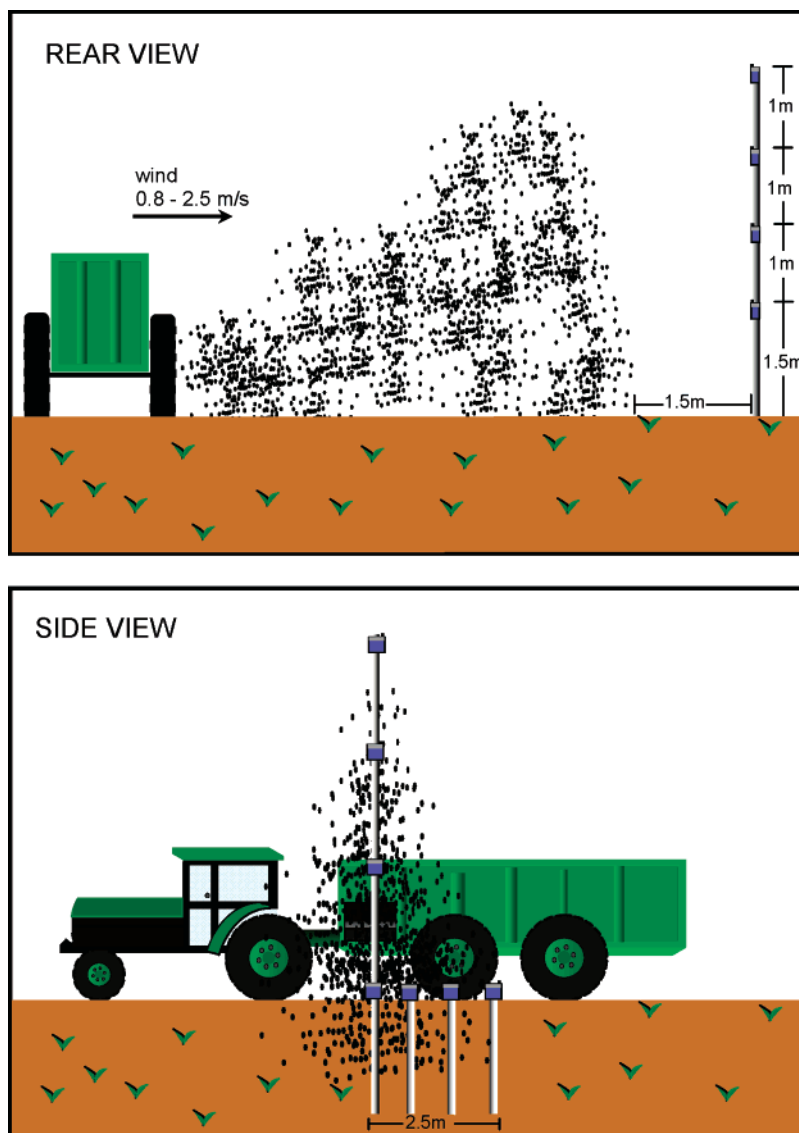
**Experimental Procedure and Sample Collection.** Field experiments were conducted in an agricultural area located southwest of Phoenix, Arizona from May 2004 to April 2005. Class B biosolids originating from one of three domestic wastewater treatment plants serving the Phoenix metropolitan area were produced by mesophilic anaerobic diges-

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**FIGURE 1.** Aerosol measurement setup during the spreading of dewatered biosolids. The figure shows  $PM_{10}$  sampler locations during concentration-weighted, cross-sectional area experiments. For source bioaerosols measurement, sampling stands were grouped (spaced 0.2–0.4 m apart) around the center of the plume and elevated to a height of 1.5 m.

tion, and mechanically dewatered (with polymer addition) to a 20–30% solids content. Dewatered biosolids were spread onto land using a side discharge slinger (ProTwin Slinger, Kuhn Knight Inc., Brodhead, MN) operated at a rate of 110 dry kg per minute.

Figure 1 demonstrates the experimental procedure for measuring aerosol source concentrations and source emission rates during biosolids land application. To eliminate dust generated by tire movement and thereby ensure that the aerosol plume was composed exclusively of biosolids-derived bioaerosols, the side discharge slinger was operated while stationary. Stationary operation, and hence aerosol experiments, were limited to approximately 1 min due to a build-up of biosolids near the exit of the slinger. Real-time  $PM_{10}$  monitoring revealed that the plume developed into a steady source within 5 s of the start of spreader operation and that the aerosol concentrations returned to ambient levels within 5 s of terminating spreader operation. Biological,  $PM_{10}$ , and metals aerosol source samplers were located 1 to 2 m downwind from the edge of the biosolids landing zone and were placed at the breathing zone height of 1.5 m. When multiple samplers were used, they were grouped (spaced 0.2–0.4 m apart) around the centerline of the source aerosol

plume. In all cases, source samplers were located such that they were directly downwind of the direction of biosolids application. Source samplers were run for the 90 s duration bracketing the 1 min time in which the biosolids spreader was operated. Control samplers for biological aerosols,  $PM_{10}$ , and airborne metals were located a minimum of 100 m upwind of any applied biosolids or spreading activities. Control aerosol sampling was performed at the same time of the day as the land application experiments and samplers were operated for a 45 min duration. To ensure a constant wind speed and direction, and control for wind aerosolization of land-applied biosolids, all experiments were performed only if wind speeds were above 0.8 m/s and below 2.5 m/s. A weather station (Weather Monitor II, Davis instrument Corp., Hayward, CA) was used in each field experiment to measure and log wind speed, wind direction, temperature, and relative humidity. All experiments were performed under slightly unstable atmospheric conditions over flat terrain.

Sterile liquid impingers (BioSampler, SKC West Inc., Fullerton, CA) were used to collect aerosol samples for total bacteria, heterotrophic bacteria plate counts (HPC), total coliforms, sulfite-reducing *Clostridia*, and endotoxin. Eight impingers were located downwind of land application and

four impingers were located at upwind control sites. Impingers were operated at a flow rate of 12.5 L/min in accordance with manufacturer specifications and flow was calibrated (Dry Cal DC-Lite, BIOS, Butler, NJ) before each experiment. The impingers were filled with 20 mL of sterile phosphate buffer saline (PBS) solution (pH 7.2, 10 mM NaPO<sub>4</sub>, 125 mM NaCl). After sampling, the impinger contents were decanted into sterile 50 mL conical tubes and the volume was recorded. Particulate matter (PM<sub>10</sub>) was measured using real-time PM<sub>10</sub> monitors (DustTrak Aerosol Monitor, model 8520, TSI Inc., St. Paul, MN) operated at a flow rate of 1.7 L/min. These monitors recorded aerosol PM<sub>10</sub> concentrations at 1 second intervals. For metal aerosol analysis, total suspended aerosol particles were collected onto a 47 mm diameter, 1 µm pore-size Teflon filter (Pall Corp., Ann Arbor, MI). The filter was attached to an open-faced support and a flow rate of 31 L/min was used during collection. Finally, aerosol samples for particle size distribution measurements were collected onto 47 mm diameter, 0.4 µm pore size polycarbonate membranes (Whatman, Florham Park, NJ). These membranes were supported by polypropylene holders (Advantec MFS, Inc., Pleasanton, CA) and loaded at flow rates ranging from 11 to 15 L/min.

Composite bulk biosolids samples were collected simultaneously with air samples by mixing biosolids collected from at least five different locations within the storage piles. Samples were placed in sterile Whirl-Pak bags (Nasco, Fort Atkinson, WI) and sealed for transportation. Solids content was determined by incubating 10 g of biosolids at 105 °C for 18 h and calculating the percent dry weight.

**Analytical Methods.** Culturable bacteria measurements (total coliforms, HPC, and sulfite-reducing *Clostridia*) for all aerosol and bulk biosolids samples were started within 2 h of collection. For bulk biosolids analysis, microorganisms were extracted from 10 g (wet weight) of biosolids by mixing with 100 mL of 0.25× Ringer's solution (38 mM NaCl, 1.4 mM KCl, 1.1 mM CaCl<sub>2</sub>, 0.6 mM NaHCO<sub>3</sub>) in accordance with previously described methods (8). For the microbial aerosol analyses, impinger samples from each experiment were pooled in order to improve the limit of detection to approximately 50 colony forming units (CFU)/m<sup>3</sup> for source aerosols and 1 CFU/m<sup>3</sup> for upwind control aerosols. The contents of two impingers were pooled for total bacteria counts, and the contents of four impingers were pooled to determine total coliforms and sulfite-reducing *Clostridia* concentrations.

Epifluorescent microscopy was used to enumerate total bacteria in accordance with previously described methods (9). Cells were stained with 4',6-diamidino-2-phenylindole (DAPI) (Pierce, Rockford, IL) at a final concentration of 20 µg/mL, filtered onto a 25 mm diameter, 0.2 µm pore-size, black polycarbonate membrane (Osmonics, Inc., Minnetonka, MN), and observed with an Olympus BX51 microscope (Olympus, Melville, NY) at 1000× magnification. HPC and total coliform plate count analysis were performed in accordance with standard methods (10). The enumeration of sulfite-reducing *Clostridia* was performed using a modified membrane filtration technique (11), where cells filtered onto a 0.22 µm Durapore membranes were anaerobically incubated at 37 °C for 48 h on an antibiotic-supplemented, egg-yolk-free tryptose–sulfite–cycloserine (TSC) agar. Endotoxin concentration measurement was conducted using the Limulus Amebocyte Lysate (LAL) Pyrochrome Kit in accordance with manufacturer instructions (ACCIUSA, Falmouth, MA). A colorimetric endpoint analysis was used to measure absorption of 405 nm light on a 96-well Vmax micro plate reader (Molecular Devices, Sunnyvale, CA). Endotoxin was quantified by comparing sample absorption to standard curves of adsorption versus concentration (Endotoxin unit (EU) per mL). Standards were re-evaluated for each reagent

batch and standard curves with an  $r^2$  below 0.97 were rejected. Endotoxin-free water and sterile impinger buffer solution were tested for blank values and all bulk biosolids and aerosols samples tested were blank corrected. Our laboratory's average coefficient of variance levels for aerosol concentration measurements are 18%, 18%, 17%, 77%, and 10% for sulfite-reducing *Clostridia*, HPC, total bacteria, endotoxin, and PM<sub>10</sub>, respectively.

To determine aerosol particle size distribution, particles collected on 0.4-µm pore size polycarbonate filters (Whatman Inc, Florham Park, NJ) were analyzed by an automated JEOL model JXA-5800 scanning electron microscope using the method described by Anderson et al. (12). Length ( $l$ ), width ( $d$ ), and area of at least 1500 particles were determined for each filter. Particle sizes were reported as the average geometric diameter,  $(l+d)/2$ . The geometric mean and standard deviation of the log normally distributed data as well as the percentage of particles under a specific size was calculated using statistical software (MINITAB 14, Minitab Inc., State College, PA). Absolute conversion between aerodynamic and geometric diameter was not possible due to the lack of information on individual particle shape and densities. The percent of total particle volume (mass) contained below a specified geometric diameter was calculated in accordance with previously published methods (13). The particle volume was estimated as the product of the SEM determined particle area and the particle width ( $d$ ).

Aerosol metal concentrations were quantified using inductively coupled plasma mass spectrometry (ICP–MS) in accordance with methods for low level aerosol particulate matter samples described by Lough et al. (14). Briefly, individual filters were digested in a microwave-assisted acid bath prior to analysis. For biosolids, 3 dry g of the sample was digested with nitric acid and hydrogen peroxide in a hot block digester and then refluxed with hydrochloric acid. The bulk biosolids and aerosol concentrations of the ten metals (As, Cd, Cr, Cu, Pb, Hg, Mo, Ni, Se, Zn) that are regulated in the U.S.EPA biosolids land application guidelines were determined (2).

**Source Emission Rate Calculation.** The aerosol emission rate  $E$  (number or mg/second) was calculated as the product of the background-corrected, source aerosol concentration  $C$  (number or mg/m<sup>3</sup>), the wind speed  $U$  (m/s), and the area  $A$  (m<sup>2</sup>) of a plane, through which the source aerosols pass, that is perpendicular to the wind direction. This area is termed the concentration-weighted, cross-sectional plume area ( $A$ ) and accounts for the spatial concentration distribution of particulate matter within this area. A novel method for determining this parameter is described. The vertical concentration profile within  $A$  was determined by placing real-time PM<sub>10</sub> monitors along the center line of  $A$  at heights of 1.5, 2.5, 3.5, and 4.5 m (Figure 1, rear view). A similar horizontal profile was determined by placing PM<sub>10</sub> monitors at a 1.5 m height at the plume center and extending 2.5 m to either side (Figure 1, side view). The vertical and horizontal boundaries of  $A$  were defined where PM<sub>10</sub> concentrations were equal to ambient PM<sub>10</sub> concentrations. Based on these horizontal and vertical concentration profiles, plume boundaries, and the assumption of a parabola shaped area (from field observations), lines of the same PM<sub>10</sub> concentrations at 0.01 mg/m<sup>3</sup> increments within the plume were plotted and shaded. These parabolic lines of equal concentrations were then used to determine the value of  $A$  (eq 1).

$$A = \text{concentration-weighted cross-sectional area} = \sum_{n=1-p}^{C_n} (A_n - A_{n-1}) \cdot \frac{C_n}{C_{1.5}} \quad (1)$$

where  $A_n$  is the area under the parabolic curve  $n$  (m<sup>2</sup>),  $n$  is



TABLE 1. Aerosol and Bulk Biosolids Concentrations and Source Emission Rates

parameter	source concentration <sup>a</sup>	upwind concentration <sup>a</sup>	bulk biosolids concentration <sup>b</sup>	aerosol source emission rate <sup>c</sup>
total bacteria (total number)	$1.8 \pm 1.0 \times 10^8$	$9.3 \pm 13.0 \times 10^6$	$4.4 \pm 3.7 \times 10^{10}$	$1.98 \pm 1.41 \times 10^9$
HPC (CFU)	$8.2 \pm 11.0 \times 10^6$	$1.0 \pm 0.6 \times 10^4$	$4.0 \pm 5.3 \times 10^8$	$9.0 \pm 11.2 \times 10^7$
total coliforms (CFU)	$380 \pm 270$	$1.3 \pm 2.8$	$1.5 \pm 1.7 \times 10^6$	$4.90 \pm 2.19 \times 10^3$
sulfite-reducing <i>Clostridia</i> (CFU)	$590 \pm 410$	$1.7 \pm 2.4$	$4.3 \pm 4.5 \times 10^5$	$6.81 \pm 3.81 \times 10^3$
endotoxin (EU)	$2.3 \pm 1.5 \times 10^3$	$3.3 \pm 2.6 \times 10^1$	$5.6 \pm 1.5 \times 10^5$	$2.1 \pm 1.8 \times 10^4$
PM <sub>10</sub> (mg)	$1.18 \pm 0.94$	0.0165	—	$10.1 \pm 8.0$
arsenic (μg)	0.060	$7.0 \times 10^{-4}$	$12 \pm 18$	$0.41 \pm 0.11$
cadmium (μg)	$3.9 \times 3.7 \times 10^{-3}$	$1.0 \times 10^{-4}$	$3.69 \pm 1.59$	$0.03 \pm 0.028$
chromium (μg)	$0.19 \pm 0.17$	0.012	$67 \pm 18$	$1.49 \pm 1.12$
copper (μg)	$0.11 \pm 0.18$	0.016	$440 \pm 10$	$0.69 \pm 1.20$
lead (μg)	$0.054 \pm 0.027$	0.024	$37 \pm 14$	$0.40 \pm 0.18$
mercury (μg)	$2.9 \pm 3.9 \times 10^{-3}$	$7 \times 10^{-4}$	$0.48 \pm 0.36$	$0.021 \pm 0.026$
molybdenum (μg)	$7.5 \pm 4.2 \times 10^{-3}$	$4 \times 10^{-4}$	$50 \pm 23$	$0.07 \pm 0.06$
nickel (μg)	$0.34 \pm 0.21$	0.017	$40 \pm 5$	$3.09 \pm 2.14$
selenium (μg)	0.34	0.016	$74 \pm 10.7$	$2.28 \pm 0.62$
zinc (μg)	$3.17 \pm 2.47$	0.16	$648 \pm 23$	$28.4 \pm 24.3$
total EPA regulated metals (μg)	$4.26 \pm 3.03$	0.25	$1372 \pm 124$	$36.9 \pm 31.8$

<sup>a</sup> Source concentration and upwind concentration per m<sup>3</sup>. <sup>b</sup> Bulk biosolids concentration per dry g. <sup>c</sup> Aerosol source emission rate per s.

the number of area concentration zones,  $p$  is the total number of area concentration zones (90),  $C_n$  is the PM<sub>10</sub> concentration (mg/m<sup>3</sup>) in area ( $A_n - A_{n-1}$ ), and  $C_{1.5}$  is PM<sub>10</sub> concentration (mg/m<sup>3</sup>) in the center of the area at 1.5 m.

All aerosol source and upwind concentrations and wind speeds were determined in four independent experiments on different days. The concentration-weighted, cross-sectional area was determined for a total of three different plumes measured on different days. To calculate emission rates for the different chemical and biological agents, the product of  $C$  and  $U$  for each independent experimental run was first calculated and then the average of this product for all experimental runs was determined. This product was multiplied times the average  $A$  to determine an overall emission rate. The  $C$  and  $U$  values used were those determined at the breathing height. Wind speed was assumed to be constant throughout the source emission area. Emission rate standard deviations were estimated by propagating errors through rate calculations in accordance with accepted methods (15).

## Results

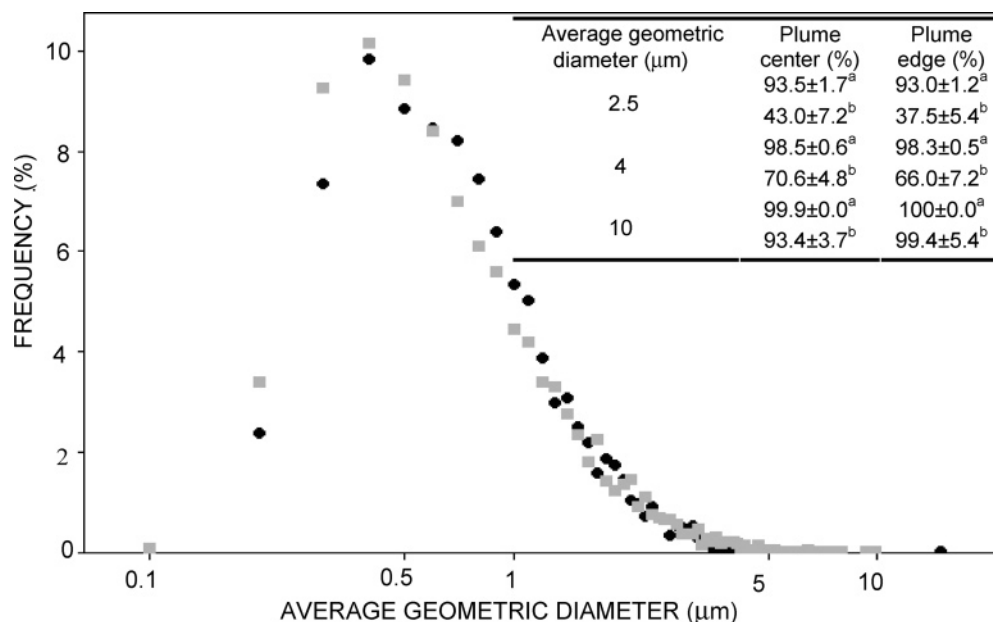
**Biosolids and Aerosol Characterization.** Table 1 lists upwind control, source aerosols, and bulk biosolids concentrations for the physical, biological, and chemical parameters considered. Real-time PM<sub>10</sub> concentration measurements at the land application source and at upwind control sites confirmed the production of aerosols during biosolids spreading. The time average PM<sub>10</sub> concentration at the aerosol source was  $1180 \pm 940 \mu\text{g}/\text{m}^3$  standard deviation and this value is more than 70 times greater than the average ambient PM<sub>10</sub> aerosol concentration measured at upwind control locations ( $p < 0.05$ ). Six geometric diameter size distribution frequencies were measured for source aerosols and include samples from three different days collected at a 1.5 m height in both the center of the plume and 2.5 m away from the center (Figure 2). Mean geometric diameter and geometric standard deviations were similar for all spreading experiments, ranging from  $1.10 \pm 0.97$  to  $1.13 \pm 0.96 \mu\text{m}$ . Greater than 98% of the particles emitted during spreading were less than  $4 \mu\text{m}$ , and an average 93% of particles were less than  $2.5 \mu\text{m}$ . Aerosols with diameters less than  $4 \mu\text{m}$  comprised the majority (70%) of the particle volume (mass).

For the microbial aerosols, statistical comparisons ( $p < 0.05$ ) confirmed that the average source aerosol concentrations of total coliforms, sulfite-reducing *Clostridia*, endotoxin, total bacteria, and HPC ( $p < 0.07$  for HPC) were higher than

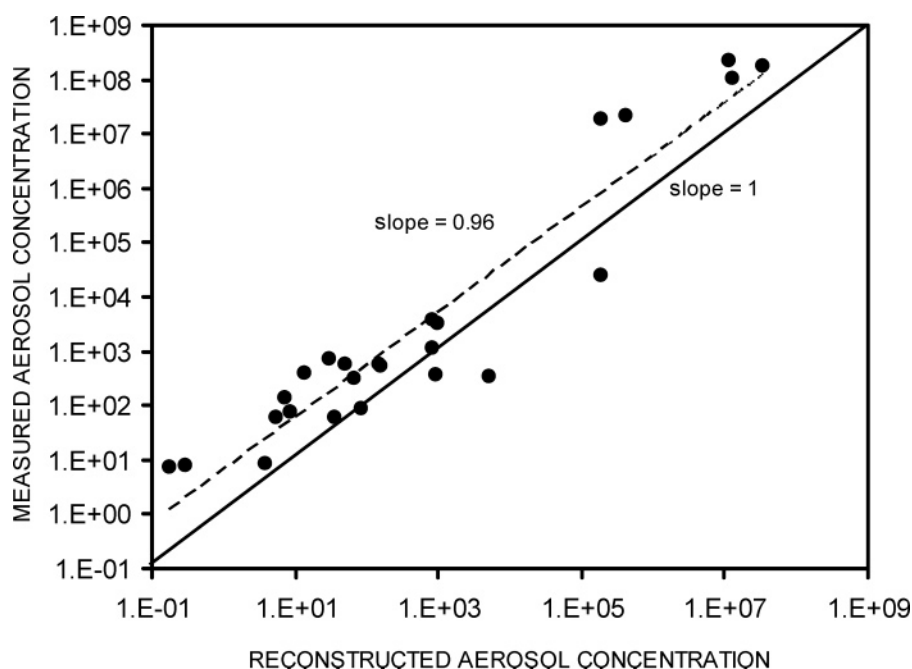
average upwind ambient aerosol concentrations. In the case of biosolids indicator microorganisms and endotoxin, this difference was substantial. Total coliforms, sulfite-reducing *Clostridia*, and endotoxin were more than 2 orders of magnitude greater at the source than at upwind locations, and upwind concentrations for indicators were not statistically different from zero. U.S.EPA regulated metals in biosolids (As, Cd, Cr, Cu, Pb, Hg, Mo, Ni, Se, Zn) were also measured in samples collected from ambient upwind aerosols and the source aerosol plume. Each of the 10 individual source aerosol metal concentrations were greater than corresponding upwind ambient concentrations, and total U.S.EPA regulated metals concentrations were an average of 12.5 times greater at the source than upwind.

Measurement of biosolids PM<sub>10</sub>, as well as bulk biosolids metals and indicator concentrations allowed for the reconstruction of source aerosol concentrations. Reconstructed aerosol concentration for a specific metal or biological agent was calculated as the product of biosolids PM<sub>10</sub> concentration (mg biosolids PM<sub>10</sub>/m<sup>3</sup>) and the concentration of the particular metal or biological agent in the bulk biosolids (ng, no., or CFU/mg bulk biosolids). The reconstructed values are independent of the measured aerosol concentration values which were determined by sampling into impingers for biological agents, and collection onto filters for metals. The two independent methods permitted a comparison between reconstructed values and measured values. Figure 3 demonstrates this comparison. The slope of a line fit to this data was 0.96 and the average reconstructed aerosol value was 21% of the average measured value. Efficiencies reported for extracting viruses and bacteria from sewage sludge range from 5 to 30%, and provide rationale into why reconstructed concentrations are less than measured concentrations (16–19). In addition, due to calibration with aerosols different from those measured in these studies, DustTrack nephelometer-style measurements should be considered only as a reasonable estimate of aerosol mass concentrations and may differ from filter-based PM<sub>10</sub> measurements (20).

**Source Emission Rates.** To determine the concentration-weighted, cross-sectional area ( $A$ ), the real-time PM<sub>10</sub> readings for each monitor in the vertical and horizontal sampling arrays were averaged over the duration of the experiment, normalized to the maximum PM<sub>10</sub> value, and either plotted versus the vertical distance along the center of  $A$  or the horizontal distance at 1.5 m height from the center to the edge of the plume. Characteristic profiles for both horizontal and vertical normalized concentration gradients in the source



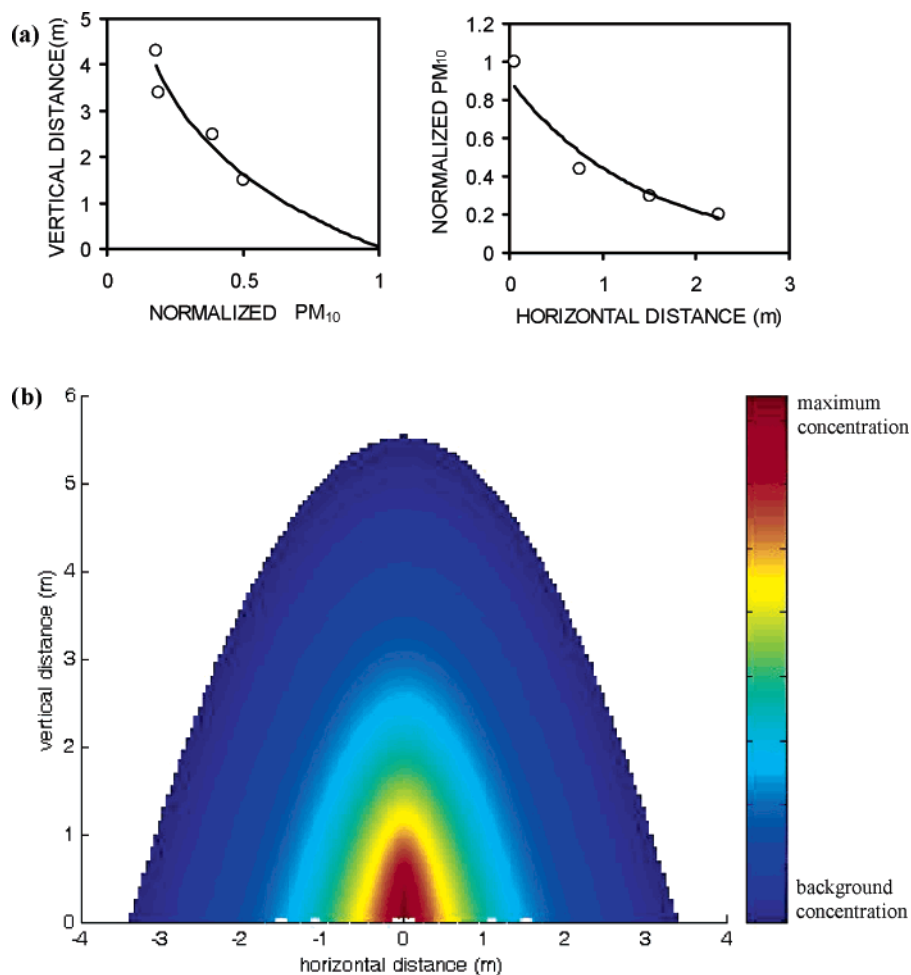
**FIGURE 2.** Characteristic profile for the geometric diameter aerosol size frequency distribution (based on size bins of 0.1 μm). Black circles represent the particle size frequency from samples taken at the center of the plume and gray squares represent the frequencies from samples taken at the edge of the plume. The table presents percentage of particles that fall below specific geometric diameter (<sup>a</sup>) as well as the percent of aerosol volume below a specific geometric diameter (<sup>b</sup>).



**FIGURE 3.** Reconstructed aerosol concentrations plotted versus measured aerosol concentrations. Data points represent concentrations of U.S.EPA regulated metals (μg/m<sup>3</sup>), endotoxin (EU/m<sup>3</sup>), total coliforms (CFU/m<sup>3</sup>), sulfite-reducing *Clostridia* (CFU/m<sup>3</sup>), HPC (CFU/m<sup>3</sup>), and total bacteria (CFU/m<sup>3</sup>). The dashed line represents a linear best fit approximation.

emission area are shown in Figure 4a. A first-order concentration decay with distance was fit for both the vertical and horizontal direction. Based on these profiles and the parabolic shape of *A* that was observed during field observation, 90 parabolic lines, each representing a 0.01 mg/m<sup>3</sup> increment in PM<sub>10</sub> concentration, were plotted and shaded (Figure 4b). This refined analysis resulted in an *A* and associated standard deviation of 7.09 ± 1.93 m<sup>2</sup>. The estimated source emission rates and standard deviations were then calculated and are presented in Table 1 for total bacteria, HPC, sulfite-reducing *Clostridia*, total coliforms, endotoxin, PM<sub>10</sub>, and biosolids regulated metals. These aerosol emission rates are independent of the wind velocity, as conservation of mass dictates

that for a steady-state emission, an increase or decrease in wind velocity results in a corresponding decrease or increase in source concentration. In addition, assuming a constant wind speed with height profile did not result in a significant over- or underestimation of the emission rates. A comparison between the calculation method describe above and an emission rate calculation which included the vertical variation of wind speed resulted in less than 10% difference between the values. For the emission areas described here, greater than 90% of the mass was below 3.5 m. In cases where source plumes extend higher into the atmosphere, variation of wind speed with height becomes substantially different from an average speed measured at the breathing height and wind



**FIGURE 4.** (a) Characteristic profile for normalized PM<sub>10</sub> concentration versus vertical and horizontal distance within the source emission cross-sectional area. Open circles represent the average normalized PM<sub>10</sub> measurements, and lines represent the best fit approximation based on first-order decay. (b) Concentration-weighted, cross-sectional area of the PM<sub>10</sub> source created during land application of dewatered biosolids via side slinging. Shades of colors represent normalized (to the center of the plume) PM<sub>10</sub> concentrations.

profile should be integrated into the emission rate calculation.

A relationship between emission rate and bulk biosolids concentration was confirmed by comparing the two parameters on a log–log plot (Figure 5). A regression between these two parameters resulted in a correlation coefficient ( $r^2$ ) of 0.96. Given the 110 dry kg/min application rate associated with the model of spreader used in this study and a PM<sub>10</sub> emission rate of  $10.1 \pm 8.0$  mg/s, the amount of biosolids aerosolized on a dry kg basis of applied biosolids was  $7.6 \pm 6.3$  mg PM<sub>10</sub>/dry kg biosolids.

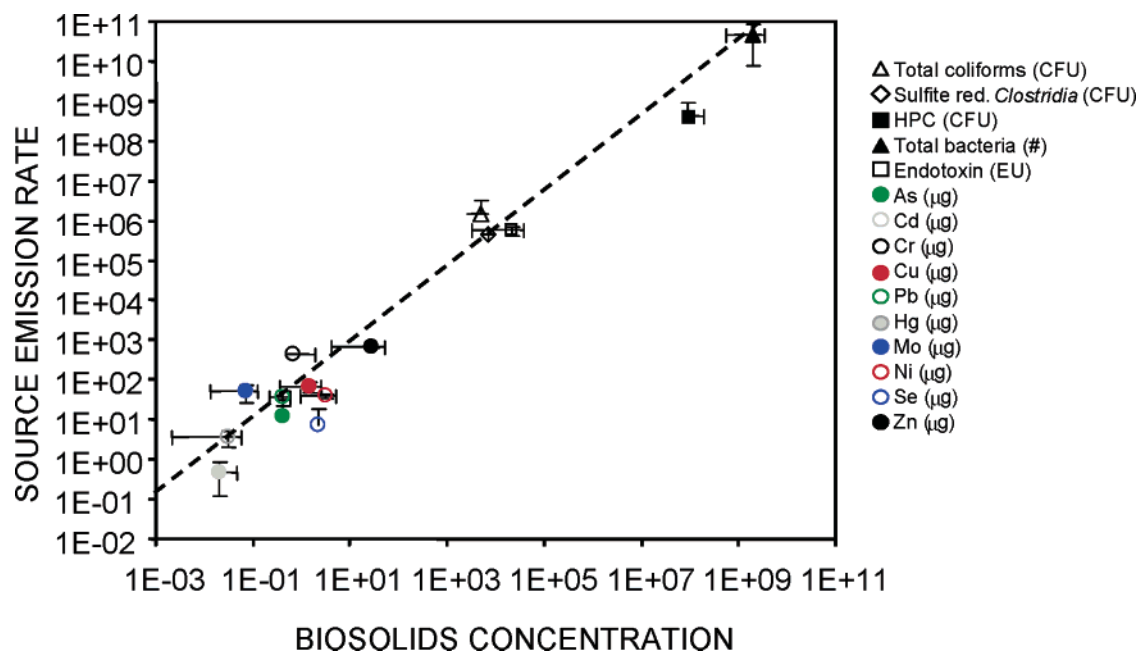
## Discussion

Aerosol concentrations of biosolids indicator microorganisms have been previously measured during the spreading of liquid and dewatered biosolids, and during the loading of dewatered biosolids (6, 7). The emissions measured herein confirm the aerosolization of dewatered biosolids during spreading. To account for the different source plume areas, the accurate comparison of emissions among different biosolids type, spreading methods, and equipment is more logically performed on the basis of an emission rate rather than source concentration. Directly measured source emission rates during biosolids land application have previously been reported only for liquid biosolids (21). Despite total coliform concentrations in bulk biosolids that were similar to the bulk concentrations in this study, source aerosol total coliforms were not detected during liquid biosolids application and the estimated liquid biosolids rates (based on total coliform

aerosol detection limits) were 35–125 times lower than the dewatered emission rates reported here. On a biosolids dry weight basis, dewatered aerosols emissions were approximately 10 mg/s versus an average 0.125 mg/s for liquid biosolids.

Size distribution, endotoxin, and metals measurements have quantitative health threshold values and provide some insight into potential biosolids-derived bioaerosol health effects in exposed populations. The percentage of emitted particles and fraction of total volume below 2.5  $\mu\text{m}$ , 4  $\mu\text{m}$ , and 10  $\mu\text{m}$  geometric diameter suggests that the majority of particles and particle volume is inhalable ( $<10$   $\mu\text{m}$  aerodynamic diameter) and may be respirable ( $<4$   $\mu\text{m}$  aerodynamic diameter). Geometric diameter, rather than aerodynamic diameter, was measured due to methodological constraints caused by the short sampling duration. Available ranges for estimating ratios of aerodynamic to geometric diameter, however, suggest that the majority of particles emitted are respirable (22). In addition to health effects, other important characteristics of particles with aerodynamic diameters below 10  $\mu\text{m}$  are slow settling velocities and consequently long residence times and travel distances in the atmosphere (13).

Although biosolids workers are near the land application zone, the use of source aerosol concentrations in estimating exposure to endotoxin and metals is less appropriate since biosolids workers would not spend substantial amounts of time directly downwind of the aerosol source. Therefore the comparison of metals and endotoxin source concentrations



**FIGURE 5.** Average aerosol emission rates produced during spreading versus average bulk-biosolids concentration. Source emission rate units are per second, whereas biosolids concentrations are units per dry gram. Error bars represent standard deviation,  $r^2 = 0.96$

with the standard threshold limit values that are provided as time-weighted average (TWA) concentrations (8 h/day, 40 h work week) would result in an overestimation of dose. Source aerosol concentrations for the ten metals considered were at least 1 order of magnitude less than corresponding TWA threshold limit values (23), indicating low aerosol metals toxicity to workers and off-site residents. In contrast, and based on recommended guidelines for endotoxin exposure (24), all source endotoxin concentrations measured were markedly greater than the 200–500 EU/m<sup>3</sup> TWA thresholds for mucus membrane irritation, and the majority of samples were above the threshold for acute bronchial constriction (1000–2000 EU/m<sup>3</sup>). While applying TWA threshold values overestimates worker exposure, it cannot yet be excluded that the real exposure time and endotoxin concentrations to which workers or nearby residents are exposed are below threshold health levels. These results corroborate previous biosolids studies that call for further investigation into endotoxin exposure (5) and help to extend endotoxin exposure data by providing an emission rate to enable aerosol transport model investigations.

In addition to the source aerosol concentrations and emission rate quantities required for use in aerosol exposure modeling, the relationships between bulk biosolids and aerosols (Figures 3 and 5) provide a broadly applicable framework for estimating aerosol concentration and emission rates from a knowledge of bulk biosolids concentration. There are distinct advantages to this fundamental partitioning approach. The first is that it allows estimation of aerosol information without the expense, difficulty, and limitations inherent in measuring bioaerosols and airborne toxins produced during biosolids land application. Commonly available aerosol sampling equipment operate at low collection rates (12.5 L/min), have efficiencies less than 100%, compromise the infectivity or culturability of microorganisms during the collection process, and do not have a well characterized collection efficiency for particles less than 0.5 µm (25). Sampling limitations are exacerbated by the fact that during biosolids application the source is mobile; requiring that collection times from stationary samplers be limited to the short time (less than 2 min) in which spreading equipment passes. These barriers may result in nondetection of important and substantial concentrations of specific toxins

or etiological agents. The approach of estimating aerosol concentrations and emissions based on PM<sub>10</sub> and bulk biosolids characterization should circumvent these limitations and move aerosol studies beyond indicator measurements by estimating specific toxic compound or pathogen aerosol concentrations based on more easily obtained PM<sub>10</sub> measurements and bulk biosolids analysis—where detection limits are much lower due to the large sample size possible. The connection between aerosol emissions and bulk biosolids toxin and pathogen concentration will allow aerosol health studies to leverage ongoing bulk biosolids toxin and pathogen monitoring results and convert them into aerosol data. Biosolids PM<sub>10</sub> concentrations also provide the inhalable particulate matter measurements that are necessary for determining the nonspecific toxicity health effects of biosolids-derived bioaerosols.

Finally, we note that while aerosol concentration reconstruction is a general concept that is broadly applicable to the variety of biosolids-derived bioaerosols that emanate from a discrete source, the relationship between source emission rates and bulk biosolids concentrations presented in Figure 5 is limited to the type of spreader used and dewatered biosolids. In this study, the ProTwin Slinger side discharge spreader was used. This particular equipment is common and the majority of land application is performed with dewatered biosolids in the 20–30% solids content range (6). When biosolids emissions are produced from a spreading scenario that cannot be accurately estimated by the common case describe here, the method described for emission rate estimation should allow for obtaining these rates through PM<sub>10</sub> and bulk biosolids measurements.

As wastewater treatment in the developed and developing world continues to move toward centralized activated sludge systems, the production of biosolids will increase accordingly. Furthermore, the much greater quantities of agricultural waste residuals that are commonly land applied, the trend of encroaching urban areas on agricultural land, and the health and nuisance complaints from off-site residents underlie the need for assessing the safety and sustainability of land application. The results presented here, while providing necessary concentration and emission rate information for assessing human exposure to biosolids during the spreading process, most importantly demonstrate a more

tractable, bulk biosolids-based approach for extending biosolids aerosol exposure assessment to different land application scenarios and to a broader range of toxins and pathogens.

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